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Incoherent interlayer transport in single-crystal films of $Nd_{2-x}Ce_xCuO_4/SrTiO_3$

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Abstract. The conductivity in $Nd_{2-x}Ce_xCuO_4/SrTiO_3$ single-crystal films with the c-axis perpendicular or parallel to the substrate plane was studied. A comparison of the results for two types of films reveals the quasi-two-dimensional character of transport processes in stoichiometric (optimally annealed) single crystal films with $0.135 \leq x \leq 0.15$ at the antiferromagnetic - superconducting phase transition.

1. Introduction

The problem of the resistivity anisotropy in the normal state of copper oxide systems has long attracted the attention of researchers. The resistivity in different directions differs not only in the magnitude but also in the character of its temperature dependence. The difference between a metallic behavior in the conducting planes ($d\rho_{ab}/dT > 0$) and an insulating behavior in the *c*- direction ($d\rho_c/dT < 0$) is a subject paid close attention by theorists [1-3], as it can give indications of the models appropriate for analyzing the normal-state resistivity of cuprates.

A strong anisotropy of the conducting properties ($\rho_c/\rho_{ab} >> 1$) when a nonmetallic conductivity along the *c* axis is combined with a metallic conductivity in the *ab* plane was repeatedly observed in underdoped and optimally doped *hole-type* HTSCs [4, 5]. This is evidence of the quasi-two-dimensionality of oxide systems that consist of highly mobile CuO_2 layers separated by buffer layers [3]. The nonmetallic character of ρ_c in most superconducting high-T_c compounds suggests an unconventional conduction mechanism between CuO_2 planes.

The cerium-doped cuprate of $Nd_{2-x}Ce_xCuO_{4+\delta}$ has a layered quasi-two-dimensional perovskite-like crystal structure [6]. As compared to other cuprate superconductors, $Nd_{2-x}Ce_xCuO_{4+\delta}$ has many unique properties that make it an attractive subject for investigations. This is a superconductor with an *electron-type* conductivity whose structure contains a single CuO_2 plane per unit cell. In optimally annealed crystals, there are no apical oxygen atoms between neighboring conducting CuO_2 planes. Therefore, $Nd_{2-x}Ce_xCuO_4$ crystals have clearly pronounced two-dimensional properties.

The $Nd_{2-x}Ce_xCuO_{4+\delta}$ compound is characterized by an ability to reversibly absorb and desorb oxygen, and its properties are very sensitive to the oxygen content. Pure Nd_2CuO_4 is dielectric, and superconductivity appears only in related solid solutions of $Nd_{2-x}Ce_xCuO_{4+\delta}$ (x > 0.13) after additional annealing in an oxygen-free atmosphere [7]. The main goal of such annealing is to remove excess nonstoichiometric oxygen (predominantly from the apical positions). This treatment reduces disorder, so electrons become delocalized and exhibit superconducting properties. After optimum doping and annealing, an $Nd_{2-x}Ce_xCuO_4$ crystal consists of a set of isolated conducting CuO_2 planes spaced 6 Å apart and is strongly anisotropic.

In bulk $Nd_{2-x}Ce_xCuO_{4+\delta}$ single crystals, a very strong anisotropy of resistivity is observed ($\rho_c / \rho_{ab} \sim 10^4$) [8-10]. However, the nonmetallic temperature dependence of $\rho_c(T)$ is quite rare. This is apparently due to the enhanced sensitivity of transport properties of the Nd system to the content of non-stoichiometric oxygen (δ) and difficulties in achieving an optimal annealing regime ($\delta \to 0$) for bulk samples. On the other hand, single-crystal $Nd_{2-x}Ce_xCuO_4/SrTiO_3$ films (up to 5000 Å thick) are well suited for different annealing procedures.

Earlier we obtained and investigated high quality $Nd_{2-x}Ce_xCuO_{4+\delta}/SrTiO_3$ single-crystal films with the c axis perpendicular [11] and parallel [12] to the substrate plane ((001) and (1 ī 0) orientations, respectively). In [13], $\rho_{ab}(T)$ and $\rho_c(T)$ dependences for $Nd_{2-x}Ce_xCuO_{4+\delta}/SrTiO_3$ single-crystal films with x = 0.12 (nonsuperconducting underdoped films), x = 0.15 (optimally doped films with a maximal T_c) and x = 0 17; 0.20 (overdoped films) at (001) and (1 ī 0) orientations have been comparatively analyzed.

Emphasis has been placed on the overdoped region, that is, in comparison with the region of optimal doping, the "damping" region of superconductivity with a gradual decrease in T_c down to $T_c \to 0$ for $x \approx 0.22$. In our Nd-system (an n-type HTSC), we have found a transition from a quasi-2D anisotropic system ($d\rho_c / dT < 0$, $d\rho_{ab} / dT > 0$) to a 3D system with metallic conductivity both in the ab- plane and in the c-axis direction ($d\rho_c / dT > 0$, $d\rho_{ab} / dT > 0$) with increased doping, x, and at higher temperatures, which is similar to that previously found in the La- system (a p-type HTSC) [5]. A conclusion about the correlation of quasi two-dimensionality of the system with the emergence of superconductivity in copper oxide compounds can be made from [5, 13].

The region of the appearance of superconductivity with increasing cerium doping (at $x \approx 0.13 \div 0.14$) thus remained unexplored. Presently, the advances in technology have allowed us to grow high-quality $Nd_{2-x}Ce_xCuO_{4+\delta}/SrTiO_3$ single-crystal films with x = 0.135 and x = 0.145 in which the c- axis was both normal ((100) films) and parallel to the substrate plane ((1 ī 0) films) in order to study the processes of charge carrier transfer in the region of the antiferromagnetic (AFM) – superconducting (SC) quantum phase transition.

The study of the properties of these samples under optimal annealing in conjunction with the optimally doped (x = 0.15) samples is the subject of this work. We are the first to experimentally observe the $\rho_c(T)$ dependences with nonmetallic behavior for samples with x = 0.135 and 0.145 that is near the threshold x values for the AFM – SC phase transition. A comparison of the results obtained for the two types of films allowed us to demonstrate the quasi-two-dimensional character of carrier transfer in them.

2. Experimental results and discussion

In table 1, we present the characteristic parameters (film thickness and the onset and completion temperatures of the superconducting transition) for both types of the investigated $Nd_{2-x}Ce_xCuO_4/SrTiO_3$ films. The results for in-plane or out-of-plane resistivities as functions of the temperature in the samples optimally annealed in vacuum with x = 0.135, 0.145 and 0.15 are shown in figure 1 (a, b).

It is seen that the normal state conductivity in the *ab*- plane is metallic with the manifestation of 2D weak-localization effects at $T \leq 70K$ in a sample with x = 0.135. The normal state out-of-plane resistivity ρ_c across the blocking layers is large with respect to the in-plane resistivity ρ_{ab} and demonstrates a non-metallic temperature dependence ($d\rho_c / dT < 0$) for all the investigated films up to 300 K.

The resistivity anisotropy coefficient is large even at room temperature ($\rho_c / \rho_{ab} \cong 10 - 10^2$) for all the investigated films (figure 2). This parameter dramatically increases with decreasing temperature and reaches the values of $\rho_c / \rho_{ab} \cong 10^3$ for the compounds with x =0.145 and 0.135 due to resistivity divergence $\rho_c(T) \sim 1/T$ at low temperatures (see figure 1).

Table 1. The characteristic parameters for both types of investigated $Nd_{2-x}Ce_xCuO_4/SrTiO_3$ films.

Type of film	x	T_c^{onset}, K	T_c, K	d, Å
	0.135	13.7	9.6	3300
(001)	0.145	15.7	10.7	2500
	0.15	23.5	22	2000
	0.135	12.8	-	4200
(1$\bar{1}$0)	0.145	15.8	6.3	4000
	0.15	21.2	7.8	1400

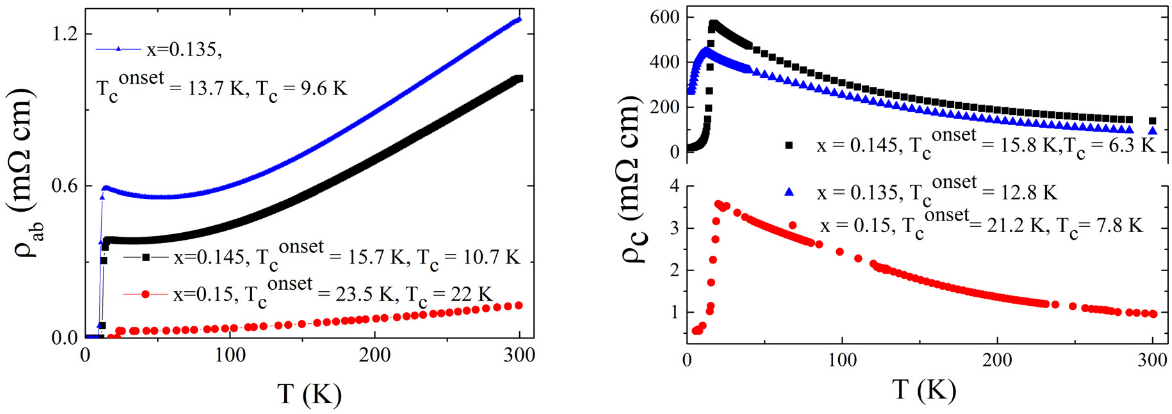

Figure 1. Temperature dependencies of in-plane (left), ρ_{ab}, and out-of-plane (right), ρ_c, and resistivity of $Nd_{2-x}Ce_xCuO_4/SrTiO_3$ films at different doping levels and optimal annealing for x = 0.135; 0.145 and 0.15.

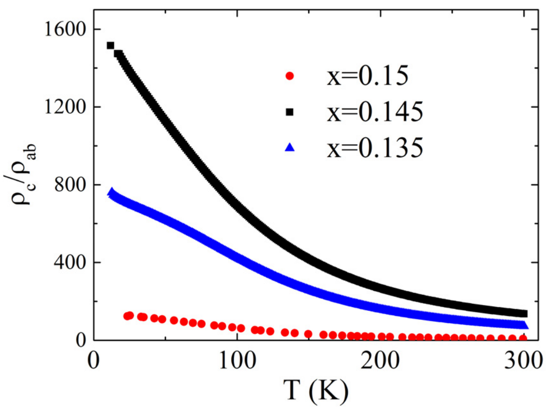

Figure 2. Temperature dependence of the resistivity anisotropy coefficient for optimally annealed $Nd_{2-x}Ce_xCuO_4/SrTiO_3$ films with various cerium contents.

A number of microscopic models for describing the deviation from a phase coherence in c-axis transport have been proposed [1-3, 14-17]. However, only the simplest one-dimensional Kronig-Penny model with its ideal periodicity (and thus the coherence) can explain the metallic nature of the interlayer conductivity.

In [14-17], the non-metallic behavior of $\rho_c(T)$ in layered oxides was attributed to *incoherent* tunneling of charge carriers in the *c*-axis direction. Incoherent transport between CuO_2 layers occurs when the probability of carrier scattering in the plane ($\hbar/\tau$) is much higher than the interlayer hopping integral t_c ($\equiv \hbar/\tau_{esc}$) between the planes. Here, τ is the carrier relaxation time in the plane, and τ_{esc} is the escape time from the given plane to the neighboring one.

If an electron experiences many collisions before moving to another plane, then the subsequent tunneling processes between the planes are uncorrelated. The interlayer conductivity is then proportional to the rate of tunneling between just two adjacent layers, and the diffusion coefficient along the layers ($D_{\|}$) and across them ($D_{\perp}$) reads (see [15], [16] and references therein):

$$D_{\|} = l^2/2\tau, \quad D_{\perp} = c^2\tau/2\tau_{esc}^2, \tag{1}$$

where l is the mean free path in the ab-plane, and c = 6 Å is the distance between neighboring CuO_2layers. Thus, we can empirically estimate the ratio of the characteristic times as:

$$\tau_{esc}/\tau = (c/l)\sqrt{\rho_c/\rho_{ab}} \,. \tag{2}$$

Using the estimates of the mean free path and the anisotropy coefficient (ρ_c/ρ_{ab}) at T < 50K, we get $\tau_{esc}/\tau \cong 10$ for x = 0.135; 0.145 and $\tau_{esc}/\tau \cong 2$ for x = 0.15. Thus, at low temperatures, the transport in the c-direction is incoherent ($\tau_{esc} >> \tau$) for underdoped Nd systems and approaches a coherent transfer ($\tau_{esc} \approx \tau$) for the optimally doped one.

We emphasize that the low-temperature anisotropy coefficient is maximal for x = 0.145 ($\rho_c/\rho_{ab} \sim 2\cdot 10^3$) and x = 0.135 ($\rho_c/\rho_{ab} \sim 10^3$) and much less for x = 0.15 ($\rho_c/\rho_{ab} \sim 10^2$) (see figure 2), in contrast to the situation in the overdoped region where the values of ρ_c/ρ_{ab} and T_c directly correlate (see figure 4 in [13]).

Using the model of a natural superlattice (when CuO_2 layers are quantum wells and Nd(Ce)O blocks serve as barriers of the effective height Δ) [17-19], we can consider the disorder that is undoubtedly inherent in this system (the chaotic impurity potential) as a cause of the incoherence in the c-axis transport.

Indeed, if the wave function of the electron is localized in the c-direction with a characteristic radius of localization r_0, which is less than the distance between adjacent CuO_2 planes, then, according to [20]:

$$t_c \sim \exp(-c/r_0)\exp(-\varepsilon_a/kT) \,, \tag{3}$$

where $r_0^{-1} = \sqrt{2m\Delta/\hbar^2}$ **,** and ε_a is the spread of electron energy in the wells due to the fluctuations of Δ values, the same as in the one-dimensional Anderson model.

The first factor in (3) (overlap integral) determines the dependence of the transition probability between the layers on the barrier height, and the second one leads to a nonmetallic temperature dependence of the conductivity at low temperatures (analogously to the conductivity within the impurity band of semiconductors [21]).

With increasing temperature, the contribution to the conductivity associated with the thermal activation of carriers across each barrier begins to play an increasingly important role [17]:

$$\sigma_{term} \sim \exp(-\Delta / kT) \tag{3}$$

and for $kT > \Delta$, there should be a transition to "metallic" conductivity. Based on the form of the $\rho_c(T)$ temperature dependence, we can estimate the effective barrier height as $\Delta > 300$ K for all the investigated samples.

3. Conclusions

We have grown high-quality (1 $\bar{1}$ 0) single-crystal $Nd_{2-x}Ce_xCuO_4/SrTiO_3$ films in which the *c* axis is parallel to the substrate plane, in order to exclude the effect of carrier transfer in the CuO_2 planes on the transport properties along the *c* direction. As a result, we were able to experimentally observe nonmetallic behavior of the normal state $\rho_c(T)$ dependence ($d\rho_c/dT < 0$) in $Nd_{2-x}Ce_xCuO_4$ samples both with $x = 0.135$; 0.145 near the AFM - SC quantum phase transition and at optimal doping with $x = 0.15$ in the SC phase.

A combination of metallic behavior of the $\rho_{ab}(T)$ dependence and nonmetallic character of the $\rho_c(T)$ dependence for stoichiometric (optimally annealed) underdoped and optimally doped NdCeCuO samples is an indication that the investigated system is quasi-two-dimensional. Specifically, two-dimensional conduction occurs along delocalized states in CuO_2 planes, and incoherent tunneling (hopping) takes place across the blocking Nd(Ce)O layers along the *c* direction.

In the present work, we found that the transport in the *c*-direction is sufficiently incoherent in the region of coexisting antiferromagnetic and superconducting ordering at $x = (0.13\div 0.14)$ and becomes more coherent in the region of SC ordering at $x = 0.15$. We assume that the fluctuations due to competition between the AFM and SC types of ordering, as well as the impurity disorder-induced spread of electron energy levels in CuO_2 quantum wells, promote the incoherent character of *c*-axis transport in the investigated structures.

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